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Measurement of chemical oxygen-iodine laser singlet oxygen generator parameter using Raman spectroscopy

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ABSTRACT

Using a doubled Nd: YAG laser as a spontaneous vibrational Raman scattering source, and a single intensified CCD array at the exit of an imaging monochromator, the Raman scattering system is used to directly measure the concentrations of the $O_2(a^1\Delta)$ and the $O_2(X^3\Sigma)$ in the chemical oxygen-iodine laser singlet oxygen generator in real time. We present the results from the tests that conducted on a 0.1-mol singlet oxygen-iodine generator. With the current reported uncertainty of the Raman cross-section, the error in the yield measurement is calculated to be less than 8%.

Keywords: chemical oxygen-iodine laser, singlet oxygen generator, Raman spectrum, yield

1. INTRODUCTION

The chemical oxygen-iodine laser is the first chemical laser based on the electronic transitions. It emits at 1315nm on the transition between the spin-orbit levels of the ground state configuration of the iodine atom. The upper level is populated by a near-resonant energy transfer from an $O_2(a^1\Delta)$ molecule to an atomic iodine atom[1-3].

$$O_2(a^1\Delta) + I(^2P_{3/2}) \rightarrow O_2(X^3\Sigma) + I^*(^2P_{1/2}) \quad (1)$$

The output power of this laser strongly depends on the yield of $O_2(a^1\Delta)$ in singlet oxygen generator[4, 5].

$$Y = \frac{[O_2(a^1\Delta)]}{[O_2(a^1\Delta)] + [O_2(X^3\Sigma)]} \quad (2)$$

To predict the COIL performance, it is very important to measure the exact values of the yield of $O_2(a^1\Delta)$ at the exit of the singlet oxygen generator. Usually, the $O_2(a^1\Delta)$ density is measured by monitoring the $O_2(a^1\Delta)$ emission at 1270nm with a calibrated detector[6, 7]. However, the detector is difficult to calibrate and the results exhibit low absolute accuracy in the yield.

Recently, the spontaneous Raman scattering technique has been developed that it can be used to directly monitor the singlet oxygen generator by measurement both the concentrations of the $O_2(a^1\Delta)$ and $O_2(X^3\Sigma)$ simultaneously using spontaneous Raman imaging[8]. It has several advantages over other methods because it allows one to monitor both the $O_2(a^1\Delta)$ and $O_2(X^3\Sigma)$ simultaneously in the same measurement volume. Because common experimental problems such as dirty windows, fluctuations in laser power, and aerosol scattering can be ratioed out, this technique greatly

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simplifies modeling and data reduction.

A Raman scattering experimental system was set up. We used it to measure the species in air and in a singlet oxygen generator. In this paper, we report the result of air and the application of Raman spectrum to measure the $O_2(a^1\Delta)$ yield at the exit of a jet-type singlet oxygen generator for COIL. The results are from the tests that conducted on a 0.1-mol singlet oxygen generator.

2. EXPERIMENT

We used the experimental setup shown in Fig. 1. A doubled Nd: YAG laser (Spectra Physics, Quanta-Ray) was used to be laser pump with a 10Hz pulse repetition rate and a 20ns pulse duration. And each pulse had a pulse energy of 400-500mJ. The output laser was elliptically polarized, and the line width was less than $1.0cm^{-1}$. The laser beam has been focused into the flow cell of singlet oxygen generator through a coated quartz window by an f/6.7 lens. The Raman scattering light which was collected perpendicularly to the propagation of the laser beam was focused throughout a Kaiser Inc. holographic $\lambda = 532$ nm notch-filter onto the slits of an f/6.5 Acton Spectrapro 500 spectrograph by a coated f/1.6 lens. The scattered Raman light was dispersed by a 1200-grooves/mm and BLZ=500nm grating and focused onto a 256×1024 ICCD array, the size of each pixel was 23μm×23μm. The ICCD chip was cooled

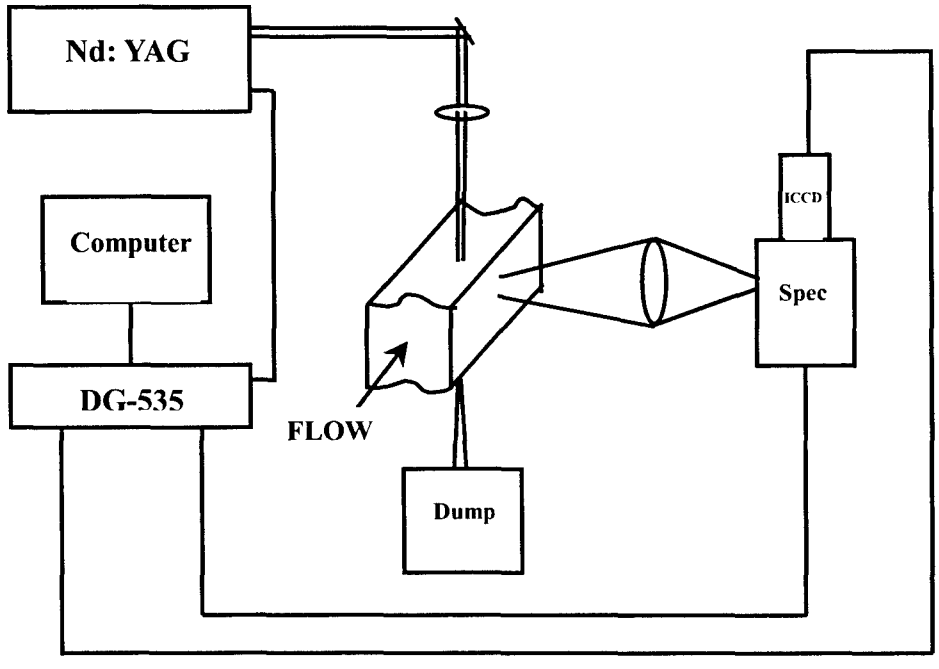


Fig.1 Schmatic illustration of experimental setup

to -30^{0}c to minimize the dark current. The DG-535 was triggered by the laser Q-Switch Sync output. At the same time, we used the computer to control DG-535 time delay so accurate that the photocathode of the image intensifier tube would be opened when the Raman scattering signal came. The detector microchannel intensifier had a gate width of 25ns. It was confirmed that this gate width could overlap each laser pulse by a 500MHz oscilloscope. The Raman cell was blacked to reduce the influence of unorderly light on optical systems.

3. RESULTS AND DISCUSSION

We used this Raman system to measure the species of air. The Raman spectrum was shown in Fig. 2. It is obtained by focusing the laser beam onto a measurement point in air. In this case, the laser output energy is about 250mJ/pulse. It is less than the energy we used in the chemical generator. The low laser output energy is needful because it can reduce the probability of optical breakdown in air. The integration time is 4 seconds. If a higher repetition rate laser were used, the same sensitivity would be obtained in shorter integration time.

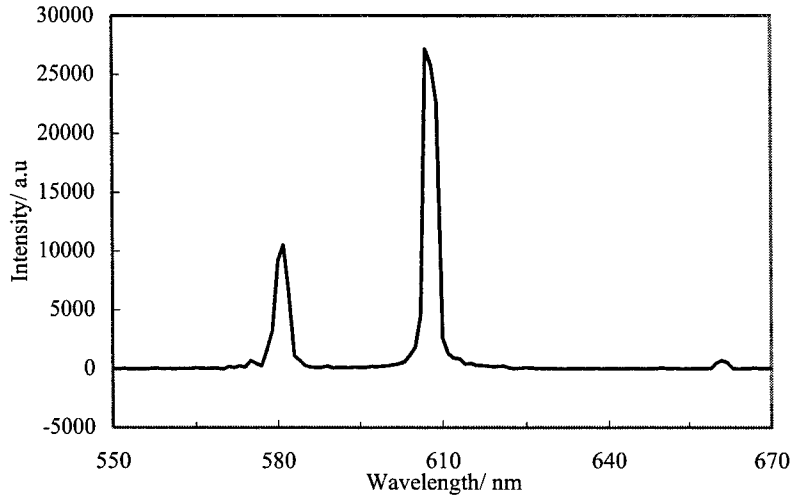


Fig. 2 The Raman spectrum of air

The typical Raman spectrums of the species in the singlet oxygen generator were shown in Fig. 3 (a) and (b). We had got each of them in 20 seconds. The Raman signal of gas in low pressure is so feeble because of the fact that Raman cross section are extremely low (~ $10^{-34}m^2sr^{-1}$) that it is very important enhancing the collecting efficiency. In addition, the Raman light should be collected at suitable angles. Which is the best angle is decided by the polarization direction of laser beam. In our tests, we collected the Raman signal perpendicularly to the propagation of the laser beam.

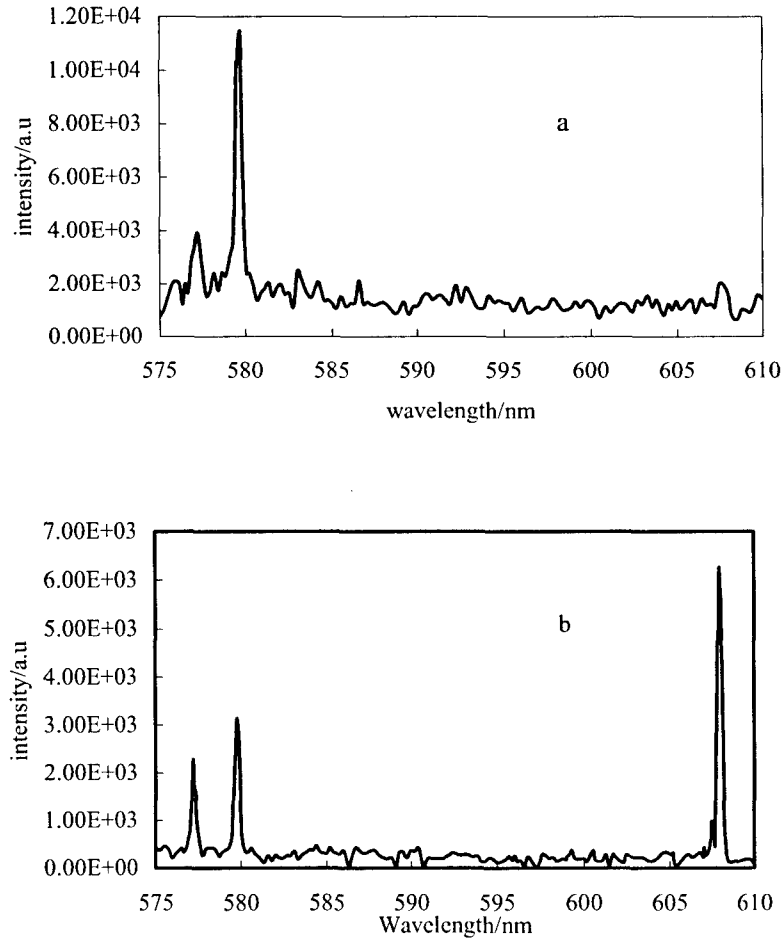
The yield can be calculated by using the expression

$$Y = \frac{[O_2(a^1\Delta)]}{[O_2(a^1\Delta)]+[O_2(X^3\Sigma)]}$$

$$= \frac{O_2(a^1\Delta)/\sigma_{O2(a)}}{O_2(a^1\Delta)/\sigma_{O2(a)} + O_2(X^3\Sigma)/\sigma_{O2(X)}}$$

$$= \frac{O_2(a^1\Delta)}{O_2(a^1\Delta) + O_2(X^3\Sigma)(\sigma_{O2(a)}/\sigma_{O2(X)})} \qquad (3)$$

where $O_2(a^1\Delta)$ and $O_2(X^3\Sigma)$ are the area under respective peaks of the $O_2(a^1\Delta)$ and $O_2(X^3\Sigma)$ Raman spectrum, σ is the Raman cross-section. $\sigma_{O2(a)}/\sigma_{O2(X)}$, the Raman cross-section ratio of the $O_2(a^1\Delta)$ and $O_2(X^3\Sigma)$ was determined by a series of experiments[9] to be 0.45.



(a) N_2: Cl_2 = 0: 106, 30Torr (b) N_2: Cl_2 = 117: 110, 18Torr

Fig. 3 The Raman spectrum of a sparger flow

The results from the tests that conducted on a 0.1-mol singlet oxygen-iodine generator had been obtained. A table of the values of a series of experiments is presented in Table 1. The differences of the optical components at the $O_2(a^1\Delta)$ and $O_2(X^3\Sigma)$ wavelengths, such as grating, detector, and notch-filter, were ignored because of the closeness of the Stokes shifts (1508 and 1580 cm^{-1}, respectively). The accuracy of the area under peak is mainly due to the signal-noise ratio. However, the yield of the $O_2(a^1\Delta)$ is in the 50% range.

Table 1 The values of a series of experiments

No.	Pressure (Torr)	N_2	Cl_2	Y (%)
1	30	0	106	39
2	35	117	106	42
3	25	117	106	45
4	25	117	106	46
5	18	117	110	54
6	9	117	104	59

It is obvious that the yield is increasing via the reducing of generator pressure in the experimental range at fixing flux, and advisable gas is desirable to be the carrier. So we should keep the singlet oxygen generator pressure lower for the sake of obtaining higher $O_2(a^1\Delta)$ yield at suitable flux. The result of experiment No. 6 is the best in Table 1. The yield is approximately 60%. With the current reported uncertainty of the Raman cross-section, the error is calculated to be less than 8%.

The sources of error in the yield measurement is as follows:

the error in the grating calibration	0.6%
the error in determining the area under the peak	2%
the error in the cross section	5%

So the total error in the yield measurement is less than 8%.

4. SUMMARY

Using a doubled Nd: YAG laser as a spontaneous vibrational Raman scattering source, and a single intensified CCD array at the exit of an imaging monochromator, the spontaneous Raman spectroscopy can be used to directly measure the singlet oxygen generator parameter of chemical oxygen-iodine lasers. The yield of $O_2(a^1\Delta)$ is in the 50% range when the flux of N_2 and Cl_2 is approximately 1:1. With the current reported uncertainty of the Raman cross-section, the error is calculated to be less than 8%. It is the most important sources of error in the yield measurement that the ratio of signal to noise is lower. It should be improved by enhancing the repetition rate, lengthening the pulse duration, improving the polarization of the laser. Reducing the unorderly light is better method to improve the ratio of signal to noise. Increased sensitivity and accuracy can be obtained with further work.

5. ACKNOWLEDGEMENTS

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